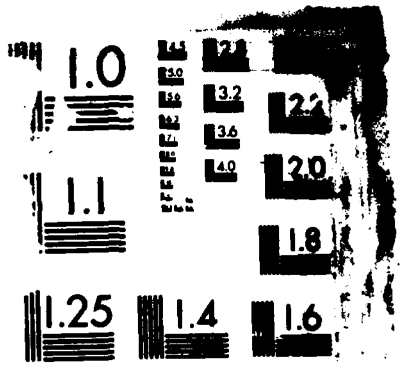
AD-A179 720 ULTRAFAST PHYSICS IN MICROSTRUCTURE AND ALLOY SYSTEMS 1/1
(U) CITY COLL NEW YORK INST FOR ULTRAFAST SPECTROSCOPY
AND LASERS R ALFANO 15 FEB 87 RF-447230
UNCLASSIFIED AFOSR-TR-87-0430 AFOSR-86-0031 F/G 9/1 NL


1.0
1.1
1.25
1.4
1.6
1.8
2.0
2.2
3.2
3.6
4.0

DTIC FILE COPY (2)

AD-A179 728

REPORT DOCUMENTATION PAGE

1a. UNCLASSIFIED

1b. RESTRICTIVE MARKINGS

2a. SECURITY CLASSIFICATION AUTHORITY

2b. DECLASSIFICATION / DOWNGRADING SCHEDULE

3. DISTRIBUTION / AVAILABILITY OF REPORT
UNRESTRICTED

4. PERFORMING ORGANIZATION REPORT NUMBER(S)
RF-447230

5. MONITORING ORGANIZATION REPORT NUMBER(S)
AFOSR-TR- 87-0430

6a. NAME OF PERFORMING ORGANIZATION
Department of Physics
The City College of N.Y.

6b. OFFICE SYMBOL *(If applicable)*

7a. NAME OF MONITORING ORGANIZATION
AFOSR/NE

DTIC ELECTE APR 24 1987 S E

6c. ADDRESS *(City, State, and ZIP Code)*
138th Street & Convent Ave.
New York, N.Y. 10031

7b. ADDRESS *(City, State, and ZIP Code)*
Building 410
Bolling AFB DC 2033-6448

8a. NAME OF FUNDING / SPONSORING ORGANIZATION
~~Dr. G. Witt~~ AFOSR

8b. OFFICE SYMBOL *(If applicable)*
NE

9. PROCUREMENT INSTRUMENT IDENTIFICATION NUMBER
AFOSR-86-0031

8c. ADDRESS *(City, State, and ZIP Code)*
Building 410
Bolling AFB DC 2033-6448

10. SOURCE OF FUNDING NUMBERS

PROGRAM ELEMENT NO.	PROJECT NO.	TASK NO.	WORK UNIT ACCESSION NO.
61102F	2305	C1	

11. TITLE *(Include Security Classification)*
Ultrafast Physics in Microstructure and Alloy Systems

12. PERSONAL AUTHOR(S)
Alfano, Robert

13a. TYPE OF REPORT
Scientific

13b. TIME COVERED
FROM 12/1/85 TO 11/30/86

14. DATE OF REPORT *(Year, Month, Day)*
2/15/87

15. PAGE COUNT
10

16. SUPPLEMENTARY NOTATION

17. COSATI CODES

FIELD	GROUP	SUB-GROUP

18. SUBJECT TERMS *(Continue on reverse if necessary and identify by block number)*
Gallium Arsenide/Gallium Aluminum Arsenide

19. ABSTRACT *(Continue on reverse if necessary and identify by block number)*

Quantum Well

During year one of AFOSR 860031, significant progress has been achieved in (I) the main thrust area of ultrafast physics in microstructures and alloys semiconductors - regarding carrier dynamics using picosecond photoluminescence kinetics of 2D ~~GaAs/GaAlAs~~ (QW) on 2 to 50 ~~psec~~ picoseconds time scale; dependence of electron temperature on well width in single ~~AlInGa/GaInAs~~ QW; determination of valence band discontinuity via optical transitions in ultrathin QW; high density electron-hole plasma energy relaxation and rapid expansion; ultrafast carrier recombination relaxation in magnetic semiconductor; and emission and relaxation dynamics in ~~0-D~~ semiconductor ~~CdSSE~~ alloy glasses; and in (II) ultrafast pulse propagation in dielectrics and semiconductors - regarding the observation of induced spectral broadening in ~~ZnSe~~ and BK-7 glass.

Cadmium Sulfur Selenide   Zinc Selenide

A total of seven papers have been published in journals - five in area (I) and two in area (II).

Details are given in the attached 10-page annual report.

Aluminum Indium Gallium/Gallium Indium Arsenide

Optical Detection

20. DISTRIBUTION / AVAILABILITY OF ABSTRACT
☒ UNCLASSIFIED/UNLIMITED ☐ SAME AS RPT. ☐ DTIC USERS

21. ABSTRACT SECURITY CLASSIFICATION
UUU

22a. NAME OF RESPONSIBLE INDIVIDUAL
Witt

22b. TELEPHONE *(Include Area Code)*
767-4931

22c. OFFICE SYMBOL
NE

During year one of AFOSR 860031, significant progress has been achieved in (I) the main thrust area of ultrafast physics in microstructures and alloys semiconductors; and in (II) ultrafast pulse propagation in dielectrics and semiconductors. A total of seven papers have been published in journals - five in area (I) and two in area (II). The following summarizes the major accomplishments made in areas (I) and (II).

In addition, during this period a SPIE conference has been organized by R. R. Alfano on the ultrafast physics in microstructures and alloys for March 1987. Two symposia were also organized for the Electrochemical Society and Laser '86 on ultrafast phenomena in condensed matter.

I. Progress in Ultrafast Physics in Microstructures and Alloys semiconductors

Great strides have been accomplished in our investigation of the **basic ultrafast physics** which occurs in quantum wells, alloys, and bulk semiconductors. Four projects have been successfully undertaken this year.

1. INVESTIGATION OF HOT CARRIERS DYNAMICS, AND RELAXATION IN 3D, 2D, 1D AND O-D SEMICONDUCTORS

The main questions we are addressing in this project are:

. the mechanisms for energy relaxation due to screening of electron-optical phonon interactions or the hot phonon bottleneck,

. the dependence of carrier capture and resonant tunneling on well thickness and dimensionality, and

. ballistic transport and rapid diffusion of carriers.

These remain important and open issues. As device's size approaches the sub-micron and nano-level, these questions become even more important. Coherent effects and breakdown of statistical based transport theories need to be re-

87 4

examined and investigated experimentally. After all hot carriers and hot phonon effects are going to play a major role no matter which semiconductor alloys are going to be used in the future ultra-small electron and optical devices. The high electron density and phonon generation will be involved in determining the operation of these ultra-small scale devices which in turn will affect their transient performance.

(a) Picosecond Photoluminescence Kinetics of 2D GaAs/GaAlAs QW

Time resolved photoluminescence (PL) kinetics on QW were performed using a 200 fs pulse from a CPM dye laser and a 4-stage amplifier system. The detector used was a streak camera with S-20 response having a state-of-the-art time resolution of 2 psec. Previous PL studies by others measured carrier temperatures kinetics beyond 20 ps. The kinetics obtained by us with finer time resolution has very interesting features. The time resolved temperature profile of the plasma does not show a monotonic decay of temperature from 2300 K at 0 ps time to about 100 K at 50 psec after excitation. A damped oscillatory behavior of carrier temperature T_e versus time is observed having a period of ~ 5 psec. This time happens to be on the order of the LO phonon lifetime in bulk GaAs. The mean temperature decay rate is much slower than the one anticipated only using existing theories of phonon bottleneck in QW. It appears the strong coupling with carriers are reheated by phonons periodically. We believe the initial decay is caused by partially screened electron-phonon polar interaction or totally screened polar interaction and relatively slow (unscreened) optical deformation potential phonon scattering (nonpolar phonons). Currently, we are investigating quantum wells of width 20 Å and 290 Å, as well as 55 Å quantum well.

Using the photoluminescence spectra of QW photoexcited by a train of high intensity subpicosecond pulses, we observed a new broad low emission feature at 4K. This emission has interesting polarization properties. The new band is

downshifted by ~ 36 meV from the electron-heavy hole 1-1 transition. This emission propagates within the plane of a Q-W and has a short recombination time of ~ 7 psec. The origin of such emission is not yet clear but may be associated with LO phonon. It is possibly connected with the stimulated emission of phonons (from the energy shift (36 meV) and lifetime (7 ps)). It may also be related to the oscillatory temperature decay rate. This work will be continued.

(b) Dependence of Electron Temperature on Well Width in the $Al_{0.48}In_{0.52}As/Ga_{0.47}In_{0.53}As$ Single-Quantum Well

Knowledge of the hot-electron behavior in semiconductor microstructures is needed to develop small and fast optoelectronic devices. It is crucial to understand the relationship between electron temperature and electron-phonon (e-p) interaction in a single quantum well (SQW) with various well widths (L_Z) and to determine how they differ from their bulk counterpart. Theoretical studies have been performed related to e-p interaction in two-dimensional (2-D) structures. Hess and his coworkers[1] predicted an enhanced e-p scattering rate over the bulk semiconductor. Ridley obtained a $1/L_Z$ relationship of 2-D e-p scattering rate by introducing a momentum conservation approximation in well direction (MCA). Leburton has shown that L_Z effect has a rather weaker influence on longitudinal optical (LO) phonon and electron interaction than Ridley's result. Although theoretical calculations predict a somewhat enhanced e-p interaction in quasi 2-D structures, the experimental results by many groups have shown a much slower hot-electron cooling rate.

We have determined the temperature of photoexcited electrons confined in a single quantum well (SQW) as a function of well width (L_Z) from L_Z = 14.5 Å to L_Z = 130.5 Å from photoluminescence spectra of a set of undoped (Al,In)As/(Ga,In)As SQW's. From electron temperature variation with L_Z it is shown that the electron

and longitudinal optical phonon scattering dominates the excess energy loss of the thermalized two-dimensional electrons in the SQW's and the scattering rate is independent of L_z within our experimental accuracy. The average energy loss rate per hot-electron was determined to be much smaller than expected. The observed variation of the steady state electron temperature with L_z indicates that the phonon scattering rate for electrons confined in SQW's does not strongly depend on the well width L_z within our experimental accuracy.

We plan to measure the picosecond kinetics of these hot carriers using our new extended S-1 streak camera (IR) with 2 ps time resolution over the range of L_z of 15 to 130 Å where emission wavelengths are 930 nm to 1400 nm.

2. Determination of Valence-band Discontinuity via Optical Transitions in Ultrathin Quantum Wells

A fundamental and long-standing problem in the study of the semiconductor microstructures is the determination of the band-edge discontinuities (ΔE_C and ΔE_v) of the heterojunctions. The intrinsic asymmetry associated with ΔE_C and ΔE_v is critical for electrons in the conduction band and holes in the valence band, respectively, to produce spectacular effects for photonic devices such as hot carrier devices, ballistic transport devices, quantum-well lasers, photodetectors, and high-speed logic elements.

We have calculated the energy separation (ΔE) between the heavy-hole and the light-hole subbands as a function of the well width for ultrathin quantum-well structures using a single-band particle-in-a-box mode. It is found that the most sensitive range for the well width (L_z) to determine accurately the valence-band discontinuity (ΔE_v) is between 15 and 80Å, whereas it is insensitive for L_z>80Å **using optical transitions** in quantum wells. A controversial issue for the determination of ΔE_v can then be resolved by measuring the ΔE's in the sensitive

well-width range. More experimental data is needed to determine ΔE_c, ΔE_v and end this controversy!

3. Photogenerated High-density Electron-hole Plasma Energy Relaxation and Rapid Expansion in CdSe

For many years probing photogeneration of an electron-hole (e-h) plasma in time provides valuable information on both the nonequilibrium and equilibrium states in terms of the interaction among elementary excitations with the environment, mainly the lattice. The carriers created from the absorption of a laser photon at energy $h\omega(h\omega > E_g)$, where E_g is the bandgap, are hot having a pseudotemperature given by $T_e \approx \frac{1}{3}(h\omega - E_g)/k_B$. Due to carrier-carrier scattering ($\approx 10^{-14}$ sec) it is assumed that the electron and hole system each have the same temperature. In our experiment, with CdSe and $h\omega \approx 2.34$ eV, the carrier pseudotemperature is ≈ 2560 K. The primary channels for energy relaxation of these hot carriers are carrier-carrier scattering, plasmon production, and phonon emission.

We have investigated the kinetics of the nonequilibrium photogenerated electron-hole plasma in CdSe using picosecond luminescence time-resolved spectroscopy. Based on the fact that the polar optical-phonon emission rate is reduced due to screening by the high density of the e-h plasma, the remaining dominant mechanism for hot-carrier cooling is the nonpolar optical-phonon emission even though CdSe is a highly polar semiconductor. It has been observed that the photogenerated carrier density is much lower than the estimated carrier density using known values of the absorption coefficient, reflectivity, and photon fluence. Rapid plasma expansion has been proposed as a possible explanation on the grounds of the observed larger spatial width of the photoluminescence relative to the

laser spatial width, moderate change of Auger recombination rate with the excitation fluence, the absence of an observed change in the Fermi level with increased excitation intensity, and earlier formation of excitons after the picosecond pulse (5 psec) excitation at a low temperature (12 K). The observed carrier density $\approx$35 psec after excitation is limited to 1×10^{19} cm^{-3} within the excitation photon fluence of 2×10^{15}-7×10^{16} photons/cm^2 at room temperature. Large values of the diffusion constant are explained in terms of a screened electron-phonon interaction. The possibility of saturation of the available states in explaining small carrier densities is eliminated by the observed faster cooling rate at low-excitation intensity and the sublinear change in the luminescence spatial width with excitation fluence.

4. Ultrafast Photoluminescence Kinetics from the Magnetic Semiconductors $CdCr_2Se_4$ Excited by Femtosecond Laser Pulse

The recombination time of photogenerated carriers in magnetic semiconductor $CdCr_2Se_4$ has been directly measured as a function of temperature using a state-of-the-art 2 ps streak camera. The relaxation kinetics of carriers has been observed to change above and below the Curie temperature 130 K. A deviation from a monotonic increase in the recombination time is measured as the temperature is lowered toward the Curie temperature. This complex behavior is attributed to the interaction between the short-range spin critical fluctuation of the magnetic ions and the spins of the carriers. The fastest measured recombination time at room temperature is 3.8 ps.

This semiconductor is ideal for ultrafast "on and off" optical switch. Work in this direction is under way.

c) Emission and Relaxation in Quasi 0-D Semiconductor Alloy CdSSe

Time resolved and steady state photoluminescence measurements were obtained from alloy CdS_xSe_{1-x} 100 Å particles 0-D embedded in glass matrix. The study of these systems allowed us to assume 1s-1s and 1p-1p transitions of the confined free electron-hole plasma is spherical QW. The measured lifetimes are short ~ 20 psec at 4K and even shorter at room temperature. Device potential for high voltage switches is yet to be explored. We plan to try these samples for switches.

II. Progress in Ultrafast Pulse Propagation in Dielectrics and Semiconductors

For many scientific and technological applications, it is important to have the capability to generate, transfer, and control a wide spectral bandwidth of ultrafast laser pulses. It is important to obtain information on the spectral distribution of an ultrafast laser pulse propagating through matter for communications[1], laser-pulse-duration control[2], and laser spectroscopy.[3] The spectral broadening of an ultrafast laser pulse as it propagates through condensed matter has been attributed to the self-phase-modulation process[3-6] and four-wave mixing.[4] Sixteen years ago, Alfano and Shapiro[4] demonstrated that an ultrafast supercontinuum pulse (USP) covering a 10 000-cm^{-1} frequency band with picosecond duration can be generated by propagating an intense picosecond laser pulse through condensed media. This phenomenon has become extremely important in the reduction of the pulse duration of 8 fs.[7] The USP has been applied to time-resolved absorption spectroscopy, nonlinear optical effects, and pulse compression. The USP could also play an important role in applications in ranging, imaging, remote sensing, communication, and other fields.

Under this grant, two major breakthroughs were achieved:

1. We made **the first observation** to our knowledge of a weak second-harmonic centered induced ultrafast supercontinuum pulse generation due to the presence of a primary intense pulse in a BK-7 glass.[8] The primary pulse at ω_1 induced the refractive change, causing a phase change and a frequency sweep of a probe pulse at $2\omega_1$. The enhancement of the bandwidth of the weak pulse at λ_2 by propagating an intense laser pulse at λ_1 in condensed media is attributed to induced phase modulation (IPM). This new effect has technological importance in communications and signal processing by permitting pulse coding in different frequency regions.

2. We made **the first observation** of induced spectral broadening (ISB) about a weak non-phase-matching second-harmonic pulse ($2\omega_0$) produced from the propagation of an intense primary (ω_0) picosecond laser pulse through a ZnSe crystal.[9] This observation can lead to the future development of **optical** information coding and modulation in different spectral regimes.

References

1. A. M. Johnson, R. H. Stolen, and W. M. Simpson, Appl. Phys. Lett. <u>44</u>, 729 (1985).

2. D. Grischkowsky and A. C. Balant, Appl. Phys. Lett. <u>41</u>, 1 (1982).

3. Ultrashort Laser Pulses, edited by S. L. Shapiro (Springer, New York, 1984).

4. R. R. Alfano and S. L. Shapiro, Phys. Rev. Lett. <u>24</u>, 584 (1970); <u>24</u>, 592 (1970); <u>24</u>, 1217 (1970).

5. J. Manassah, M. Mustafa, R. Alfano, and P. P. Ho, IEEE J. Quantum Electron. <u>22</u>, 197 (1986).

6. J. Manassah, M. Mustafa, R. R. Alfano, and P. P. Ho, Phys. Lett. <u>113A</u>, 242 (1985).

7. W. H. Knox, R. L. Fork, M. C. Downer, R. H. Stolen, and C. V. Shank, Appl. Phys. Lett. 46, 1120 (1985).

8. R. R. Alfano, Q. X. Li, T. Jimbo, J. T. Manassah, and P. P. Ho, Optics Letters 11, 626 (1986).

9. R. R. Alfano, Q. Z. Wang, T. Jimbo, and P. P. Ho, Phys. Rev. A35, 459 (1987).

III. Plans for the Future

Area I - Microstructure

1. The time resolved photoluminescence and absorption spectroscopy will be used to determine capture times and tunnelling times between and among the Q-W and barriers.

2. Direct measurement of free carrier spin relaxation in 2-D systems using picosecond spectroscopy and its dependence on L_z.

3. Systematic study of fast diffusion and ballistic transport of carriers in 2-D, 1-D and 0-D system.

4. Carrier dynamics in AlInAs/GaInAs single QW by time resolved PL kinetics using S-1 camera.

5. Hot hole dynamics will be investigated in QW and bulk materials using ps IR spectroscopy (3 to 5 μ).

6. Construct and test aan optical switch with the magnetic semiconductor $CdCrSe_4$ at room temperature and low temperature. Also, CdSSe glass switches will be tested.

Area II - Pulse Propagation

1. Measure of time dependence of IBM and ISB processes in glass and ZnSe.

IV. Publications During Period

1. K. Shum, P. P. Ho, R. R. Alfano, D. F. Welch, G. W. Wicks, and L. F. Eastman, Dependence of Electron Temperature on Well Width in the $Al_{0.48}In_{0.52}As/Ga_{0.47}In_{0.53}As$ Single-Quantum Well, IEEE J. Quant. Electr. QE-22,1811-1815 (1986).

2. K. Shum, P. P. Ho, and R. R. Alfano, Determination of Valence-band Discontinuity via Optical Transitions in Ultrathin Quantum Wells, Phys. Rev. B33, 7259-7262 (1986).

3. M. R. Junnarkar and R. R. Alfano, Photogenerated High-density Electron-hole Plasma Energy Relaxation and Experimental Evidence for Rapid Expansion of the Electron-hole Plasma in CdSe, Phys. Rev. B34, 7045-7062 (1986).

4. P. P. Ho, W. Lam, A. Katz, S. S. Yao, R. R. Alfano, Temperature Dependence of the Ultrafast Photoluminescence Kinetics from the Magnetic Semiconductors $CdCr_2Se_4$ Excited by Femtosecond Laser Pulse, IEEE J. Quant. Electr. QE-22, 205-208 (1986).

5. M. R. Junnarkar and R. R. Alfano, Spin Dephasing Kinetics of Free Carriers in Alloy Semimagnetic Semiconductors $Cd_{1-x}Mn_xSe$ by One and Two Photon Excitation, in **Ultrafast Phenomena V**, ed. G. R. Fleming and A. E. Siegman, pp. 251-253, Springer Verlag (1986).

6. R. R. Alfano, Q. X. Li, T. Jimbo, J. T. Manassah, P. P. Ho, Induced Spectral Broadening of a Weak Picosecond Pulse in Glass Produced by an Intense Picosecond Pulse, Optics Lett. 11, 626-628 (1986).

7. R. R. Alfano, Q. Z. Wang, T. Jimbo, P. P. Ho, R. N. Bhargava, B. J. Fitzpatrick, Induced Spectral Broadening about a Second Harmonic Generated by an Intense Primary Ultrashort Laser Pulse in ZnSe Crystaly, Phys. Rev. A35, Rapid Communications 459-462 (1987).

END
5_87
DTIC